Characteristics of particle number and mass concentrations in residential houses in Brisbane, Australia

Lidia Morawska[1], Congrong He[1], Jane Hitchins[1], Kerrie Mengersen[2] and Dale Gilbert[3]

[1]International Laboratory of Air Quality and Health, Queensland University of Technology, Brisbane, QLD 4001, Australia
[2]School of Mathematical and Physical Sciences, The University of Newcastle, University Drive, Callaghan, NSW 2308, Australia
[3]Built Environment Research Unit, Queensland Department of Public Works and Housing, Brisbane, QLD 4001, Australia

Abstract

Diurnal variation of indoor submicrometer (0.007 – 0.808μm) particle number and particle mass (approximation of $PM_{2.5}$) concentrations was investigated in fifteen houses in a residential suburb of Brisbane during winter in 1999. Continuous monitoring for more than 48 hours was conducted using a condensation particle counter (CPC) and a photometer (DustTrak) in the kitchen of each house, and the residents kept diaries of the activities conducted. In addition, data from a central monitoring station was used to investigate indoor/outdoor relationships. The results show that there were clear diurnal variations in both particle number and approximation of $PM_{2.5}$ concentrations, for all the investigated houses. The pattern of diurnal variations varied from house to house, however, there was always a close relationship between the ~~variations~~ concentration and human indoor activities. The average number and mass concentrations during indoor activities were $(18.2\pm3.9)\times10^3$ particles cm^{-3} and (15.5±7.9) μg m^{-3} ~~in the approximation of $PM_{2.5}$ range and~~ respectively, and under non-activity conditions, $(12.4\pm2.7)\times10^3$ particles cm^{-3} (11.1±2.6) μg m^{-3}, respectively. In general, there was a poor correlation between mass

and number concentrations and the correlation coefficients were highly variable from day to day and from house to house. This implies that ~~no~~ conclusions cannot be drawn about either one of the number or mass concentration ~~one of these~~ characteristics of indoor particles, ~~(number or mass concentration)~~ based on measurement of the other. The study also showed that it is unlikely that particle concentrations indoors could be ~~accountably~~ represented by measurements conducted at a fixed monitoring station ~~monitoring~~ due to the large impact of indoor and local sources.

Keywords: indoor particles, indoor air quality, particle number concentration, submicrometer particles, indoor $PM_{2.5}$

1. INTRODUCTION

Understanding of human exposure ~~of~~ to indoor particles is important to enable exposure control and reduction. The earlier studies on indoor particles provided information and background knowledge mainly on mass concentrations and on coarse particles, such as total suspended particles (TSP), PM_{10} and some also on $PM_{3.5}$ fractions (mass concentration of particle matter with aerodynamic diameter smaller than~~t~~ 10 µm, 3.5 µm, respectively). This was due to the limited understanding of the relationship between particle size and the health effects they cause, as well as to instrumental limitations. More recently, a few studies on the indoor fine particle mass and number concentrations have been published, however, the available information on fine and ultra fine particles indoors, both in terms of mass ~~concentration~~ and number concentration, and the relationship between mass and number concentration, is still relatively limited.

Another clear weakness in the databases provided by previous studies on indoor particles is the lack of information about the spatial and temporal or short-term variations of fine particle concentration indoors. One of the main reasons for this is that traditional filter sampling methods require sampling times in the range between 24 ~~or~~ to 48 hours to collect a sufficient ~~amount of~~ mass of particles. Such sampling methods are thus unsuitable for conducting real-time particle concentration studies. On the other hand, the TEOM (Tapered Element Oscillating Microbalance monitor) instrument commonly used for real-time outdoor measurements of particle mass (Soutar et.al. 1999), is not the most convenient instrument to be used indoors, due to its relatively large size, making it a nuisance for the occupants. Its ~~and~~ high flow rate~~,~~ also affect~~sing~~ the indoor ~~conditions of~~ air exchange rates and thus pollutant concentrations. In consequence, it is difficult to accurately estimate the risk due to particle exposure by using the existing databases because indoor particle levels may exhibit significant short-term variability (e.g.; Brauer et al., 1999), which can potentially ~~as discussed above, can~~ have significant health implications.

There are a few studies in the literature that have simultaneously and continually measured indoor fine particle number and mass concentration for periods up to 48 hours (Long et al., 2000; Wigzell et al., 2000). An emerging conclusion from these studies is that the pattern of diurnal variation of indoor fine particle concentrations is different from the pattern of diurnal variation of outdoor fine particle concentrations, and that indoor activities may significantly affect fine particle concentration indoors. Continuous real time monitoring of indoor fine particles can provide the ~~needed~~ required information on the impact of the sources with time-varying intensity on indoor fine particle concentrations and thus lead to improvement in the understanding and assessment of exposure.

In order to address this need and to better understand exposure to indoor particles, the objectives of this study were to: 1) investigate the diurnal variation of particle number concentration and particle mass concentration (approximation of $PM_{2.5}$) in 15 residential houses; 2) ~~analyse the~~ quantify the impact of indoor activities on indoor particle concentration levels based on real-time measurements and time-activity information; and 3) compare the indoor particle concentrations with ambient particle concentrations measured at a central monitoring station. The present study is part of a larger program, some of the outcomes of which have already been published or submitted for publication (Morawska et al., 2001; Hargreaves, 2002; Ayoko et al., 2002)

2. EXPERIMENTAL METHODS

2.1. *Sampling Site and Houses*

A residential suburb on the south-eastern side of Brisbane, about 10km from the city centre and of reasonably flat topography was chosen as the measurement site. Fourteen houses in this suburb and one additional house for comparison, located to the east of the city were identified for the study. Ideally, houses for the study should be identified through a random selection process. ~~Unfortunately~~ however it was not feasible to use such a process for this study for two reasons~~.~~; firstly, ~~T~~the houses had to be selected from one area to remove ~~uncertainness~~ uncertainties related to additional factors, such as variation in topography, socioeconomic status, etc., and secondly, ~~one hand, and on the other hand~~ the studies were highly intrusive to the occupants, therefore acceptance rate was very low. The houses investigated in this study differed in age (from 2 to100 years), construction material (timber, brick), stove type (electric, gas) and design (high set, low set). High set means that the house is elevated above ground on timber or brick stumps and low set, that the house is built

directly on the ground. House design and material characteristics have an effect on the air exchange rate, but as this parameter is not included in the analyses presented in this paper, detailed house characteristics are not provided. In general, air exchange rate was estimated to range for all the houses in this study from 0.3 to 6.4 for normal ventilation conditions, and from 0.1 and 2.0, for minium ventilation condition (all windows and doors closed). More details about the houses can be found in Morawska et al., 2001; Hargreaves, et al., 2002 and Ayoko et al., 2002. The majority of the houses were occupied by non-smokers, with Houses 5, 12 and 17 the only exceptions. However, on occasions there were ~~these houses occasionally~~ visitors to ~~these~~ some houses who smoked. The occupants of Houses 5 and 17 however never smoked inside. All the houses were naturally ventilated.

2.2. *Instrumentation*

The real-time total number concentrations of submicrometer particles in the range from 0.007 to 0.808 μm were measured using the TSI Model 3022A Condensation Particle Counter (CPC) (TSI Incorporated, St. Paul, MN, USA), with an inlet impactor (0.0508 cm nozzle) and aerosol flow rate of 0.3 L/min.

The TSI Model 8520 DustTrak aerosol monitor (TSI Incorporated, St. Paul, MN, USA) with a 2.5 μm inlet was used to measure the real-time approximation of $PM_{2.5}$ concentration. It should be noted that the DustTrak operates based on a light scattering technique where the amount of scattered light is proportional to the volume concentration of the aerosol. The approximation of $PM_{2.5}$ values obtained in this study using this instrument are not actual gravimetric values, as the instrument was not calibrated for each specific aerosol studied. It was, however, compared against TEOM (Tapered Element Oscillating Microbalance monitor) for a general type of indoor

aerosol, from which correction factors were derived enabling better estimation of $PM_{2.5}$ than ~~this~~ that based on the manufacturer's calibration.

These instruments were chosen as the most suitable for an indoor study because of: 1) the low flow rates required ~~and~~ thus having little impact on particle concentrations indoors;~~,~~ 2) their quiet operation ~~which means less noise and thus fewer nuisances~~ causing less intrusion to the occupants;~~,~~ and 3) the short sampling times of 10 and 30 seconds respectively for the CPC and DustTrak, and thus providing~~sion of~~ real-time data.

Comparison of the indoor approximation of $PM_{2.5}$ and particle number concentrations with respective outdoor concentrations was conducted using the data collected from the Air Monitoring and Research Station (AMRS). The Station, located in the Brisbane CBD is part of the South East Queensland monitoring network, and is operated by the Queensland Environmental Protection Agency. In addition to other parameters, PM_{10} concentrations are monitored in the Station using a TEOM (50ºC R&P 1400a) as well as particle number concentration using the TSI model 3934 Scanning Particle Sizer (SMPS) operating in the size range from 0.016 to 0.7 μm. Unfortunately $PM_{2.5}$ is not monitored in the Station.

2.3. *Measurements*

Particle number concentrations and the approximation of $PM_{2.5}$ concentrations were measured simultaneously for more than 48 hours in the kitchen of all the houses during March and July 1999, which is autumn/winter time in Brisbane. The CPC and DustTrak were placed side-by-side and positioned on average two metres from the stove in the kitchen. These measurements followed a few hours of intensive

monitoring of particle number concentration and size distribution, approximation of $PM_{2.5}$, fungi, VOC's and NO_x concentrations under different ventilation conditions and with all the known indoor particle sources switched off. Some of the results of these investigations have been published elsewhere (Morawska et al., 2001). The occupants of the houses were required to fill in a diary, noting the time and duration of any activity conducted during the time of the measurements.

2.4. *Data processing, correction and analysis*

All statistical analyses (correlation, regression, t-test, one-way ANOVA) were conducted using a statistical analysis software package, SPSS for Windows version 10 (SPSS Inc.). Because the distribution of the concentration measures was not normal, a robust analysis in which the maximum and minimum quartiles were trimmed was employed. In addition, nonparametric tests were undertaken to confirm the parametric results. That is, the corresponding nonparametric tests led to the same conclusions of significance/nonsignificance as the parametric tests.

Corrections for DustTrak and CPC

Since the DustTrak operates on the principle of light scattering, its response is highly dependent on the size distribution and refractive index of the sampled aerosol. This instrument was factory-calibrated with Arizona dust particles and it was reported that for finer aerosols such as commonly encountered in indoor air, the instrument's response can be significantly higher than the true $PM_{2.5}$ value (Ramachandran et al., 2000). In order to obtain values closer to true $PM_{2.5}$ values from the data collected by the DustTrak, an additional experiment was conducted under laboratory conditions to compare the DustTrak indoor readings with the readings of a Tapered Element

Oscillating Microbalance (TEOM) monitor (50ºC R&P 1400a with a URG $PM_{2.5}$ cyclone inlet). Ambient particles were used in the experiment. Based on the results of this additional experiment, the following linear regression equation was obtained:

$$PM_{2.5(TEOM)} = 0.394\ PM_{2.5(DustTrak)} + 4.450; \quad (\text{with } R^2 = 0.83) \qquad (1)$$

All the approximation of $PM_{2.5}$ data collected by the DustTrak in this study were corrected using this equation. The non-zero intercept of this equation is most likely the result of low sensitivity of the DustTrak to particles in the lower submicrometer range. While their contribution to $PM_{2.5}$ mass is normally not very high, yet it has a non-zero value. It should be noted that the values of $PM_{2.5}$ derived using this equation are still approximations of the true value, because different indoor activities may require somewhat different factors in the equation. Yet, as discussed above, it is expected that the values derived using equation 1 will differ from true values not more than 17%, which is much less than it would be relying only on the factory calibration, when the difference sometimes is up to a factor of a few.

Recently, Yanosky et al., (2002) conducted a comparison study in an indoor environment (but without regular occupants) in which a DustTrak and a US EPA designated Federal Reference Method (FRM) $PM_{2.5}$ sampler (gravimetric method), the BGI, Inc. PQ200, were assessed ~~the comparability of~~to compare the two sampling methods. They found that the 24-h average DustTrak levels are well correlated with FRM levels (R^2 = 0.859) but show significant proportional bias. The correction equation obtained from their study was as follows:

$$PM_{2.5(FRM)} = 0.33\ PM_{2.5(DustTrak)} + 2.25; \quad (\text{with } R^2 = 0.859) \qquad (1A)$$

~~Comparison~~ Examining Equations 1 ~~with Equation~~and 1A, it ~~could find~~can be seen that the parameters in the two equations are comparable.

The CPC used in this study for the indoor measurements and the SMPS used for outdoor measurements had, due to instrumental constraints, slightly different ranges of operation. Due to this as well as to the fact that they are different systems, it was expected that they would yield somewhat different results when sampling the same aerosol. In order to provide a more accurate relationship between indoor and outdoor particle number concentrations, a comparison between the two instruments was conducted at the AMRS. Based on the results of this comparison, the following linear regression equation was obtained:

$$C_{SMPS} = 0.4213\ C_{CPC} + 3019;\ (\text{with } R^2 = 0.67) \qquad (2)$$

where C_{SMPS} and C_{CPC} are the total number concentrations measured by the SMPS and the CPC, respectively. When comparing indoor and outdoor particle number concentrations, the CPC data were adjusted using this equation.

It is acknowledged that transformation through the above equations induces uncertainty that is not appropriately accounted for in the ensuing analyses. Although the transformed values are 'best estimates' in that they are expected values and unbiased, any tests of significance made on their basis should be interpreted with caution.

Processing of particle data

The results from particle measurements were classified as follows and the average concentrations for each class were calculated:

- Based on the real-time concentration data, the total 24h average indoor concentrations were calculated for each house.

- Based on activity records, the situations indoors were classified into one of two categories: non-activity conditions and activity conditions, meaning lack or occurrence of occupant activities inside the house. As the next step, situations indoors under the non-activity conditions were further classified into one of two categories: daytime (about 06:30 - 20:00) and night-time (about 20:00 - 06:30). Under the activity conditions, the indoor situations were further classified into morning activities, day activities and evening activities. Average particle concentrations were calculated for each of these situations for each house. In addition, average concentrations were calculated for all non-activity situations that occurred during each 24 h period.
- Based on the real-time outdoor concentration data, outdoor total 24h average concentrations were calculated. Further, the outdoor concentrations were also classified into daytime (about 06:30 - 20:00) and night-time (about 20:00 - 06:30).

Finally, in order to compare the indoor and outdoor particle mass concentrations, the outdoor PM_{10} were adjusted to $PM_{2.5}$ using the ratio of $PM_{2.5}/PM_{10} = 0.61$. This value was provided by the Environmental Protection Agency (Neale D. and Wainwright D. 2000) based on the data from the monitoring stations in Brisbane for 1999, the year in which this project was conducted.

3. RESULTS

3.1 *Diurnal variation*

Real-time particle concentration data combined with the human activity information allow the sources of particle diurnal variations to be analysed. The results show that there were clear diurnal variations (one-way ANOVA, $p<0.005$) of particle number

concentration and the approximation of $PM_{2.5}$ concentration for all the investigated houses with the pattern of the variations differing from house to house. In general, however, the particle concentrations in the morning (06:00 - 09:00) and evening (17:00 - 20:00) were higher than the concentrations during midnight to early morning, and daytime (09:00 - 17:00). This trend in particle concentrations correlates well with the pattern of human indoor activity where most cooking occurs in the evening or in the morning.

Typical diurnal variations of particle number and approximation of $PM_{2.5}$ concentrations are presented in Figures 1 and 2~~(Congrong – some of the activities don't appear to have lines indicating their time – or are mis-aligned in figure 2)~~. The figures show how human activities, especially cooking, can significantly affect the indoor particle concentration. There is a sharp increase in concentration at the start of each indoor activity and a slow decrease after the activity ceased. It can be seen from Figure 1 that both particle number and mass concentrations were very low in the absence of human activities (for example: CPC about $1.5 \sim 2.5 \times 10^{3}$ particles cm^{-3} and approximation of $PM_{2.5}$ about $6 \sim 8$ μg m^{-3} for the period from 16:00 hour, 9 July to 19:00 hour, 10 July). However, particle concentrations were significantly higher during cooking activities (up to 100 times higher than the background level) and often remained high for a long period of time of up to 14 hours, as presented in Figure 1 for the period from 19:00 hour, 11 July to 9:00 hour, 12 July. A more detailed discussion of the concentration levels and their variation for different times and activities is provided below.

3.2 *Characteristics of variation in particle number concentrations*

Table 1 present a summary of concentrations for each situation and for each house under the classification developed for this study. The results presented in Table 1 show that the total 24h average particle number concentration ~~(Congrong – the total 24h average seems as though it is under the "Activity" column – it doesn't have the heading "Total 24h". Maybe you could insert some vertical lines to distinguish each section ie Non activity, activity, total, Min, max, ratio max/min, and ratio NA24/24 – and see if it looks clearer?)~~is clearly higher (t-test, $p < 0.001$) than non-activity 24h average concentration for all houses with the ratio of the former to the latter ranging from 1.23 to 2.31, and with an average value of 1.49 (±0.3). Furthermore, the average ratio of the 24h indoor to the 24h outdoor concentration (1.35±0.88) is higher (t-test, $p = 0.001$) than that of the non-activity 24h indoor concentration to the outdoor 24h concentration (1.05±0.72). (See Table 1)

It can be seen from Table 1 that the daytime outdoor concentrations had an average of $(12.9\pm7.2)\times10^{3}$) particles cm^{-3} and were always higher (t-test, $p = 0.005$) than the night-time concentrations, which averaged $(8.1\pm5.0) \times 10^{3}$ particles cm^{-3}. There were no significant differences between the indoor and outdoor daytime concentrations under non-activity situations. The ratios of daytime indoor to outdoor concentrations under non-activity conditions averaged 0.93±0.67 and were statistically lower (t-test, $p = 0.024$) than the ratios of night-time concentrations, which averaged 1.33±0.74. This implies that the daytime indoor concentrations under these conditions were mainly influenced by outdoor concentrations, while the night-time indoor concentrations were affected by the activities that took place in the houses in the evening. During the winter, the residents of these houses normally keep windows open during the day and closed at night. This behaviour results in reduction of the air exchange rate at night, thus reducing the amount of outside air entering the house at

night-time and reducing removal of particles generated indoors during evening activities.

The minimum particle number concentration ranged from 1.3×10^3 to 7.8×10^3 particles cm^{-3} with an average of $(4.2\pm1.9)\times10^3$ particles cm^{-3}. The times when the concentrations were at a minimum were between about ~~11:00pm~~23:00 h and 08:~~00am~~00 h and between ~~02:00pm~~14:00 h and ~~06:00pm~~18:00 h in the absence of human activities. The maximum number concentration ranged from 1.12×10^5 to 8.17×10^5 particles cm^{-3} with ~~the~~ an average of $(2.69\pm2.38)\times10^5$ particles cm^{-3}. The maximum number concentrations were observed between about 06:30~~am~~ h and 08:50~~am~~ h and between about ~~4:30pm~~16:30 h and ~~8:00pm~~20:00 h during cooking times. The maximum to minimum ratio of particle concentrations ranged from 15 to 238, with an average of 73±62. These large standard deviations in the minimum, maximum and maximum/minimum ratio results from the high~~ly~~ variability of the particle number concentration indoors. These results not only indicate that the diurnal variation of particle number concentration was house-specific and highly dependent on the resident's activities and lifestyle, but also quantify the ranges of concentrations and variation in the concentrations between the houses.

3.3 *Characteristics of variation in the approximation of $PM_{2.5}$*

Table 1 summarises also measured indoor and recalculated outdoor approximations in $PM_{2.5}$ concentrations. Under the non-activity conditions, the daytime and night-time approximation of $PM_{2.5}$ concentrations varied somewhat from house to house, with a daytime average for all the houses of 10.6±2.6 μg m^{-3} and night-time average of 11.7±3.0 μg m^{-3}. Unlike particle number concentrations, there were no significant differences between the daytime and night-time approximation of $PM_{2.5}$

concentrations for most houses. The ratios of daytime approximation of $PM_{2.5}$ indoor concentrations for non-activity conditions to outdoor concentrations, varied only slightly from house to house, with ~~the~~ an average of 1.06±0.2. This result is consistent with the findings of Morawska *et al.*, (2001) that the ratios of indoor to outdoor approximation of $PM_{2.5}$ concentration for conditions ~~where~~ of no indoor sources ~~operated~~ were close to one for most houses during the daytime (1.01 and 1.08 for normal and minimum ventilation conditions, respectively). The ratios of night-time indoor to outdoor concentrations varied from house to house and were greater than one for all houses, except House 1, with an average of 1.71±0.64. The outdoor night-time $PM_{2.5}$ concentrations averaged at 7.3±2.8 μg m^{-3} and were significantly lower (t-test, $p < 0.001$) than the daytime outdoor concentration, most likely due to the decrease in human activities outdoors during the night. Nine of the fifteen houses investigated in this study displayed higher night-time than daytime indoor approximation of $PM_{2.5}$ concentrations. Further analysis revealed that the residents of the nine houses (except for~~one,~~ House 14) normally prepare their main meal in the evening. Although the cooking ended in the evening, the indoor approximation of $PM_{2.5}$ concentrations remained higher for prolonged periods of time during the night due to the low air exchange rate at night during winter. In summary, the ratios of night-time indoor to outdoor $PM_{2.5}$ concentrations are greater than one and are significantly greater (t-test, $p = 0.001$) than the ratios of the daytime concentrations.

Both 24h non-activity average and the total indoor approximation of $PM_{2.5}$ concentrations varied from house to house, but the latter showed a more significant variation with an average of 11.1±2.6 μg m^{-3} for non-activity conditions and 15.5±7.9 μg m^{-3} for total concentrations. In the absence of an Australian $PM_{2.5}$ standard, these average concentrations can be compared to the US EPA $PM_{2.5}$ 24h and annual

standards of 65 and 15 μg m^{-3}, respectively. While the 24h standard was not exceeded in any house in this study, the concentration levels are close to the value of the annual standard.

The ratios of 24h total to non-activity indoor approximation of $PM_{2.5}$ concentrations were greater than one for all houses with an average of 1.36±0.49. For most of the houses, however, the ratios were not much higher than one, with only a few houses where the ratios were significantly elevated (t-test, $p < 0.001$) ~~elevated~~.

The minimum approximation of $PM_{2.5}$ concentrations indoors normally occurred over the period from ~~1:00am~~01:00 h to ~~7:00am~~07:00 h and were in the range from 5.2 μg m^{-3} to 8.8 μg m^{-3} with an average of (7.3±1.2) μg m^{-3}. The maximum concentrations ranged from 16 μg m^{-3} to 2.8×10^{3} μg m^{-3} with an average of $(5.35\pm9.09)\times10^{2}$ μg m^{-3}. The maximum concentrations normally coincided with cooking activities and were observed between ~~8:00am~~08:00 h and ~~2:00pm~~14:00 h and between ~~5:30pm~~17:30 h and ~~8:00pm~~20:00 h. The ratio of maximum to minimum approximation of $PM_{2.5}$ concentrations ranged from 1.8 to over 2200. As for the number concentrations discussed above, these large standard deviations in the minimum, maximum and in the ratios of maximum to minimum concentrations result~~s~~ from the high~~ly~~ variability of the approximation of $PM_{2.5}$ concentration indoors. These results not only indicate that the diurnal variation of approximation of $PM_{2.5}$ concentration was house-specific and highly dependent on the resident's activities and lifestyle, but also quantify the ranges of concentrations and variation in the concentrations between the houses.

The ratios of 24h total indoor to outdoor concentrations were greater than one for all houses except House 1, with the average for all the houses of 1.72±1.00. The ratios of 24h non-activity indoor to outdoor concentrations were ~~L~~lower than these ~~were the~~

~~ratios of 24h non-activity indoor to outdoor concentrations~~ with an average of 1.27±0.28. Comparison of the ratios not only demonstrates that the indoor activities increased the concentration of the indoor approximation of $PM_{2.5}$ which became higher than the outdoor concentration, but also quantify the extent of this increase.

3.4 *Comparison of number to mass concentration*

The real-time paired data points of indoor particle number concentrations and approximation of $PM_{2.5}$ concentrations were analysed to identify in the first instance the existence of linear correlations. The results showed that correlation coefficients (r) were highly variable from day to day, and from house to house. For example, the correlation coefficient for House 14, varied from 0.39 (first day) to 0.69 (second day) and then 0.59 (third day). The correlation coefficient for House 4 changed from 0.24 (first day) to 0.67 (second day). The average correlation coefficient for each house ranged from 0.1 (House 6) to 0.73 (House 3), with an overall average value of 0.48±0.27. There were significant correlations ($p < 0.01$) between the indoor particle number concentrations and approximation of $PM_{2.5}$ concentrations on some days, but not on other days. Therefore in summary, there were no clear linear correlations between approximation of $PM_{2.5}$ and particle number concentration in the long-term~~,~~ ~~which~~ This could be due to a number of factors, the most important of them being that different sources could be the main contributors to particle number and approximation of $PM_{2.5}$ concentrations.

The results of further correlation analysis showed there were some other types of relationship (power, logarithmic and exponential) in some cases. However, in general~~ly~~, there was no ~~any~~ long-term, consisten~~tcy~~ relationship between approximation of $PM_{2.5}$ concentration and number concentration. This implies that no

conclusions can be ~~drawn~~made ~~on~~ about one of these characteristics of indoor particles (number or mass concentration), based on ~~the~~ a measurement of the other.

4. DISCUSSION

Figure 2 shows that both particle number concentrations and approximation of $PM_{2.5}$ concentrations clearly increased during the time when the residents were smoking in the room. The peak concentrations were up to two times, and five times higher than the background values, for number and mass concentrations, respectively. Many previous studies showed that cigarette smoking significantly affects indoor particle concentration. For example, Brauer et al., (2000) found that the peak concentrations of $PM_{2.5}$ were up to 4.5 times higher than the background values in a bar during cigarette smoking. They also report the range of $PM_{3.5}$ concentration during cigarette smoking in a residential living room was 20 ~ 100 μg m^{-3}. Their results are comparable with the findings in this study (32 ~ 50 μg m^{-3} for $PM_{2.5}$ concentration during cigarette smoking).

Investigations of diurnal variation in outdoor particle mass concentrations showed that the high outdoor concentrations were normally observed during the traffic peak hours. This result is consistent with previous findings that the patterns of outdoor diurnal variations were significantly affected by the diurnal variations in vehicle emissions and meteorological conditions in Brisbane city (Morawska et al., 1999; Jamriska et al., 1999).

When comparing indoor and outdoor diurnal variations, it was found that the patterns were quite different when there were significant indoor activities occurring. This finding is expected and consistent with, for example, the results reported by Patterson and Eatough (2000) who measured the diurnal variation of indoor and outdoor fine

particle number and mass concentration ($PM_{2.5}$) in a school using a CNC and a TEOM. Their results showed clear diurnal variations in indoor particle concentration. However, the authors found a low correlation between indoor $PM_{2.5}$ concentrations and outdoor $PM_{2.5}$ concentrations because of the influence of student activities on the indoor concentrations.

A previous results from the same large study (Morawska et al., 2001), showed that the ratios of indoor to outdoor particle number concentrations were on average below one during the day when no indoor sources were in operation (0.89 and 0.78 under normal and minimum ventilation, respectively). Those findings are consistent with the results presented here where the average ratio of daytime non-activity concentrations indoor and outdoor concentrations was 0.93±0.67. Unlike studies of particle mass concentration indoors, there are only a few studies reporting~~ed~~ on indoor particle number concentrations. Li *et al.,* (1993) investigated real-time indoor submicrometer particle concentrations in the size range: 0.017-0.886 μm for 24 hours. Their results showed that the concentration varied from 1.4×10^4 to 1.5×10^5 particles cm^{-3}. This is comparable with the results of this study and not only supports the finding that peak values indoors occur during significant indoor activities~~,~~ such as cooking, but also provides ~~the assessment~~an estimate for ~~of~~ the ~~high~~height of these peaks.

Several different measurement methods were used for monitoring of indoor $PM_{2.5}$ concentrations in the studies reported in the literature, therefore comparisons of the results from these studies should be ~~taken~~ made with caution. Wiener, et al. (1990) performed a long-term study of $PM_{2.5}$ concentrations in nine homes in the USA and the results showed that the average 24 hours $PM_{2.5}$ concentration was (36.3±2.6) μg m^{-3}. Recently, Lachenmyer and Hidy (2000) examined the average mass

concentration in 10 houses in Birmingham, AL, USA using filtration and gravimetric mass techniques. Their data showed that the average indoor 48 hours $PM_{2.5}$ concentration was (16.1±5.7) $\mu g.m^{-3}$ in summer, and (11.2 ± 5.4) $\mu g.m^{-3}$ in winter. In a study conducted over two weeks in 28 homes in an urban area of Huddersfield, U.K., Kingham, et al., (2000) measured $PM_{2.5}$ concentrations using active pump samplers running for 24h in each home. The mean values were (17.81±12.21) $\mu g.m^{-3}$ for houses located within 50 m of the main road and (19.52±13.58) $\mu g.m^{-3}$ for houses at a distance greater than 50 m of from the main road, respectively. In Perth, Australia, Stratico and Dingle (1996) using a respirable dust cyclone found that the average 24 hours $PM_{2.5}$ concentration of 48 houses was 34.4 ± 20.3 $\mu g.m^{-3}$. Monn et al., (1997) measured indoor $PM_{2.5}$ concentrations in Switzerland using Teflon filters. The sampling period was from 48 to 72 hours. The average daily $PM_{2.5}$ concentration was found to be 18.3 $\mu g.m^{-3}$ for non-activity situations and 26.0 $\mu g.m^{-3}$ for activity situations.

The results of the current study are comparable with the findings of Lachenmyer and Hidy (2000) and Kingham, *et. al*., (2000), are somewhat lower than those of Monn et al (1997) and are significantly lower than those of Wiener, *et. al*., (1990) and Stratico and Dingle (1996). The large~~r~~ difference between the results of these~~s~~ studies may be contributed to by the different measurement methods~~,~~ which were used for monitoring of indoor $PM_{2.5}$ concentrations ~~in these studies~~, as well as differences in the indoor/outdoor environments investigated.

Comparing the indoor particle concentrations it was apparent that a low concentration of approximation of $PM_{2.5}$ does not necessarily imply a low particle number concentration and vice versa (for example Houses 4 and 12 and House 6, respectively). These findings suggest that the daily correlations between particle

number concentration and $PM_{2.5}$ concentration are complex and variable, and are highly affected by the number, type and strength of indoor activities and outdoor sources. It was clear that the average number concentrations including indoor activity conditions were higher than those for non-activity conditions for all houses, but not all the houses showed the same trend in relation to mass concentration (approximation of $PM_{2.5}$). This result suggests that number concentration is a better particle characteristic than $PM_{2.5}$ ~~of~~ to determine the contribution from indoor sources to the total concentration (the most important of which are combustion sources). ~~than $PM_{2.5}$ concentration.~~

5. CONCLUSIONS

In this study significant short-term variations in particle number concentrations and approximation of $PM_{2.5}$ concentrations ~~was~~ were found in fifteen houses in Brisbane, Australia. The results show that there were clear diurnal variations in both particle number and approximation of $PM_{2.5}$ concentrations, for all the investigated houses. The pattern of diurnal variations varied from house to house, however, there was always a close relation between the variations and human indoor activities. The average number and mass concentrations during indoor activities were $(18.2\pm3.9)\times10^3$ particles cm^{-3} and (15.5 ± 7.9) μg m^{-3}, respectively, and ~~in the approximation of $PM_{2.5}$ range and respectively~~ under non-activity conditions, $(12.4\pm2.7)\times10^3$ particles cm^{-3} (11.1 ± 2.6) μg m^{-3}, respectively. The results of this study support the findings of previous studies that the major sources of particles indoors are cooking and environmental tobacco smoke (for smoking indoors). In general, there was a poor correlation between mass and number concentrations and the correlation coefficients

were highly variable from day to day, and from house to house. This implies that no conclusions can not be drawn about either one of the number or mass concentration these characteristics of indoor particles (number or mass concentration) based on measurement of the other.

The aAnother conclusion from this study is that due to the significant differences between the indoor and outdoor diurnal variations it would be very difficult to accurately estimate indoor particle mass or number concentrations by using the outdoor concentrations solely.

The limitations of the study were the relatively short time of measurements in each house, the relatively small number of houses investigated and using only an approximation of $PM_{2.5}$. measured, yet its The strengths was were in the provision of real-time data with high resolution, enabling estimation of minimum and maximum in concentration as well as the time series concentrations.

ACKNOWLEDGMENTS

This project was supported by the Built Environment Research Unit, Queensland Department of Public Works, and the Australian Research Council, through SPIRT grant No. C69804416. The assistance of Keith Eigeland and Chris Greenaway is gratefully acknowledged. Members of the QUT International Laboratory of Air Quality and Health, in particular, Sandhya Parappukkaran, Milan Jamriska, Stephen Thomas and Graham Johnson, are appreciated for their discussions and assistance during the course of this study. The authors would like to express their special gratitude to the owners and occupants of the houses for their help and assistance with this project, without which this project could not have been conducted.

REFERENCES

Ayoko, G.A., Morawska, L., Hitchins, J., Gilbert, D., Christensen, E., 2002. Sources and concentration levels of volatile organic compounds in residential houses in Brisbane, Australia. Submitted for publication

Brauer M. Hirtle R. Lang B. Ott W. 2000. Assessment of indoor fine aerosol contributions from environmental tobacco smoke and cooking with a portable nephelometer. *Journal of Exposure Analysis & Environmental Epidemiology*. 10(2):136-144.

Brauer, M., Hirtle, R.D., Hall, A.C., Yip, T.R., 1999. Monitoring personal fine particle exposure with a particle counter. *Journal of Exposure Analysis & Environmental Epidemiology*. 9(3):228-236.

Burke, J.M., Zufall, M.J., Ozkaynak H. 2001. A population exposure model for particulate matter: case study results for PM2.5 in Philadelphia, PA. *Journal of Exposure Analysis & Environmental Epidemiology.* 11(6):470-489.

Hargreaves M., Parappukkaran S., Morawska L., Hitchins J., He C., Gilbert D., (2002) A pilot investigation into associations between indoor airborne fungal and non-biological particle concentrations in residential houses in Brisbane, Australia. Submitted for publication

Jamriska, M., Thomas, S., Morawska, L., Clark, B., 1999. Relation between indoor and outdoor exposure to fine paticulates near a busy arterial road. *Indoor Air*, *9*, 75-84.

Kingham, S., Briggs, D., Elliott, P., Fischer, P., Lebret, E., 2000. Spatial variations in the concentrations of traffic-related pollutants in indoor and outdoor air in Huddersfield, England. *Atmospheric Environment*. 34(6):905-916.

Lachenmyer, C., Hidy, G.M., 2000. Urban measurements of outdoor-indoor PM25 concentrations and personal exposure in the deep south. Part I. Pilot study of mass concentrations for nonsmoking subjects. Aerosol Science & Technology. 32(1):34-51.

Li CS. Lin WH. and Jenq FT. 1993. Characterization of outdoor submicron particles and selected combustion sources of indoor particles. *Atmospheric Environment Part B-Urban Atmosphere*. 27(4):413-424.

Long, C.M., Suh, H.H., Koutrakis, P., 2000. Characterization of indoor particle sources using continuous mass and size monitors. *Journal of the Air & Waste Management Association*. 50(7), 1236-1250.

Monn, C., Fuchs A., Hogger, D., Junker, M., Kogelschatz, D., Roth, N., Wanner, H.U., 1997. Particulate matter less than 10 mu-m (PM10) and fine particles less than 2.5 mu-m (PM2.5) - relationships between indoor, outdoor and personal concentrations. *Science of the Total Environment.* 208(1-2), 15-21.

Morawska, L, Thomas, S, Gilbert, D, Greenaway, C, Rijinders, E., 1999. A study of the horizontal and vertical profile of submicrometer particles in relation to a busy road. *Atmospheric Environment* 33:1261-1274

Morawska, L., He, C., Hitchins, J., Gilbert, D., Parappukkaran, S., 2001. The relationship between indoor and outdoor airborne particles in the residential environment. *Atmospheric Environment*. 35 (20), 3463-3473.

Morawska, L, Thomas, S, Gilbert, D, Greenaway, C, Rijinders, E., 1999. A study of the horizontal and vertical profile of submicrometer particles in relation to a busy road. *Atmospheric Environment* 33:1261-1274

Neale, D., Wainwright, D., 2000. Ambient air quality monitoring in Queensland 1999 annual summary and trend report. Queensland Government Environmental Protection Agancy.

Patterson, E., Eatough, D.J., 2000. Indoor/outdoor relationships for ambient PM2.5 and associated pollutants:Epidemiological implications in Lindon, Utah. *Journal of the Air & Waste Management Association*. 50(1), 103-110.

Soutar, A., Watt, M., Cherrie, J.W., Seaton, A., 1999. Comparison between a personal PM10 sampling head and the tapered element oscillating microbalance (TEOM) system. *Atmospheric Environment*. 33(27):4373-4377.

Stratico, A., Dingle, P., 1996. Background fine particulate levels in Australia homes. In: *Proceedings of the 7th International conference on Indoor Air Quality and Climate*, Vol. 1. 591-594.

Wiener, R.W., Wallace, L.A., Pahl, D., Pellizzari, E.D., Whitaker, D., Spengler, J., Özkaynak, H., 1990. Rewiew of the Particle TEAM 9-home field study. In Proceedings of the EPA-A&WMA Symposium on Measurement of Toxic Chemicals. Air & Waster Management Association (VIP-17), Pittsburgh, PA., 452-460.

Wigzell, E., Kendall, M., Nieuwenhuijsen, M.J., 2000. The spatial and temporal variation of particulate matter within the home. *Journal of Exposure Analysis & Environmental Epidemiology*. 10(3):307-314.

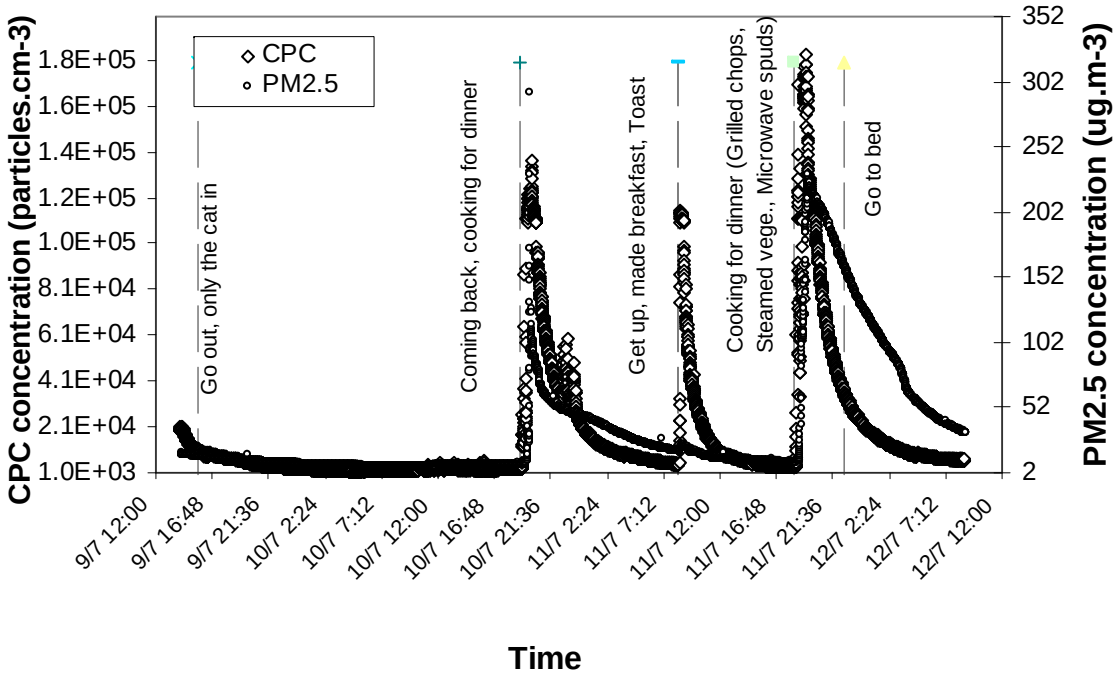
Yanosky J.D., Williams P.L. and MacIntosh D.L., (2002) A comparison of two direct-reading aerosol monitors with the federal reference method for $PM_{2.5}$ in indoor air, *Atmospheric Environment*. 36(1): *107-113.*

Table 1. Summary of the diurnal variation of indoor and ambient particle number concentrations (particle number concentration: particles cm^{-3} ×1000) and the fine particle mass concentrations ($PM_{2.5}$: μg m^{-3}), as well as the indoor and outdoor concentration ratios.

	Non-Activity			**Activity**				Min	Max	Ratio	Ratio	**Day**	**Night**	**Total**	**Ratio**	**Ratio**	**Ratio**	**Ratio**
	Day	Night	24h	Morning	Day	Evening	24h			Max/Min	A24h/NA24h			**24h**	**DI/DO**	**NI/NO**	**NAI24h/O24h**	**I24h/O24h**
Particle Number*																		
Average	**12.8**	**12.0**	**12.4**	**44.4**	**22.3**	**41.1**	**18.2**	**4.2**	**268.6**	**73.4**	**1.49**	**12.9**	**8.1**	**10.9**	**0.93**	**1.33**	**1.05**	**1.35**
S.D	3.3	3.4	2.7	18.6	8.3	18.4	3.9	1.9	237.0	61.7	0.30	7.2	5.0	6.1	0.67	0.74	0.72	0.88
Max	17.4	20.8	19.1	85.9	35.4	77.4	27.2	7.8	816.6	238	2.31	24.5	20.0	22.1	2.72	2.70	2.88	3.52
Min	7.6	8.5	9.0	24.7	8.5	19.8	13.6	1.3	112.1	15.0	1.23	3.5	2.7	2.8	0.27	0.59	0.35	0.52
Particle Mass																		
Average	**10.6**	**11.7**	**11.1**	**18.5**	**11.8**	**47.3**	**15.5**	**7.3**	**535.4**	**77.3**	**1.36**	**10.0**	**7.3**	**8.9**	**1.06**	**1.71**	**1.27**	**1.72**
S.D	2.6	3.0	2.6	17.3	4.6	80.4	7.9	1.2	909.9	134	0.49	3.2	2.8	3.1	0.20	0.64	0.28	1.00
Max	16.6	19.1	17.5	78.6	26.7	320.7	36.9	8.8	2842	473	2.71	16.3	12.0	15.3	1.32	3.27	1.79	4.86
Min	7.7	8.0	7.9	8.2	8.0	10.3	8.0	5.2	15.9	1.81	1.02	6.5	4.2	5.6	0.59	0.88	0.71	0.75

Min: Minium; Max: Maximum; 24h: 24 hours average; NA24h: Non-Activity 24 hours average; A24h: total 24 hours average; 24h: 24 hours average; DI/DO: Day Indoor/Day Outdoor; NI/NO: Night Indoor/Night Outdoor; NAI24h: Non-Activity Indoor 24 hours average; I24h: total Indoor 24 hours average; O24h: Outdoor total 24 hours average.

*Due to instrument malefaction data on particle number concentration was not available for one of the houses, therefore average values for number concentration were calculated for fourteen, not fifteen houses.


CPC
PM2.5
Go out, only the cat in
Coming back, cooking for dinner
Get up, made breakfast, Toast
Cooking for dinner (Grilled chops, Steamed vege., Microwave spuds)
Go to bed
CPC concentration (particles.cm-3)
PM2.5 concentration (ug.m-3)
1.8E+05
1.6E+05
1.4E+05
1.2E+05
1.0E+05
8.1E+04
6.1E+04
4.1E+04
2.1E+04
1.0E+03
352
302
252
202
152
102
52
2
9/7 12:00
9/7 16:48
9/7 21:36
10/7 2:24
10/7 7:12
10/7 12:00
10/7 16:48
10/7 21:36
11/7 2:24
11/7 7:12
11/7 12:00
11/7 16:48
11/7 21:36
12/7 2:24
12/7 7:12
12/7 12:00
Time

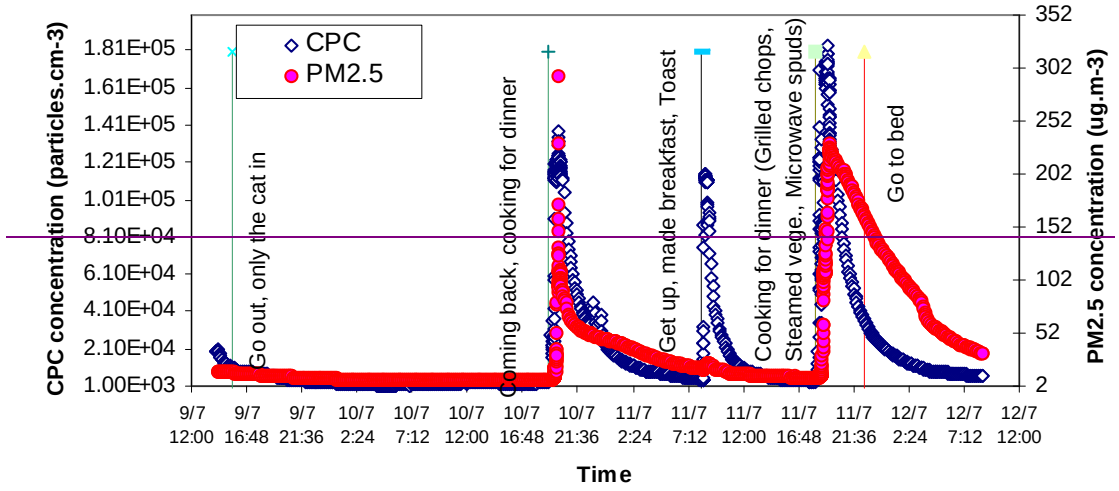
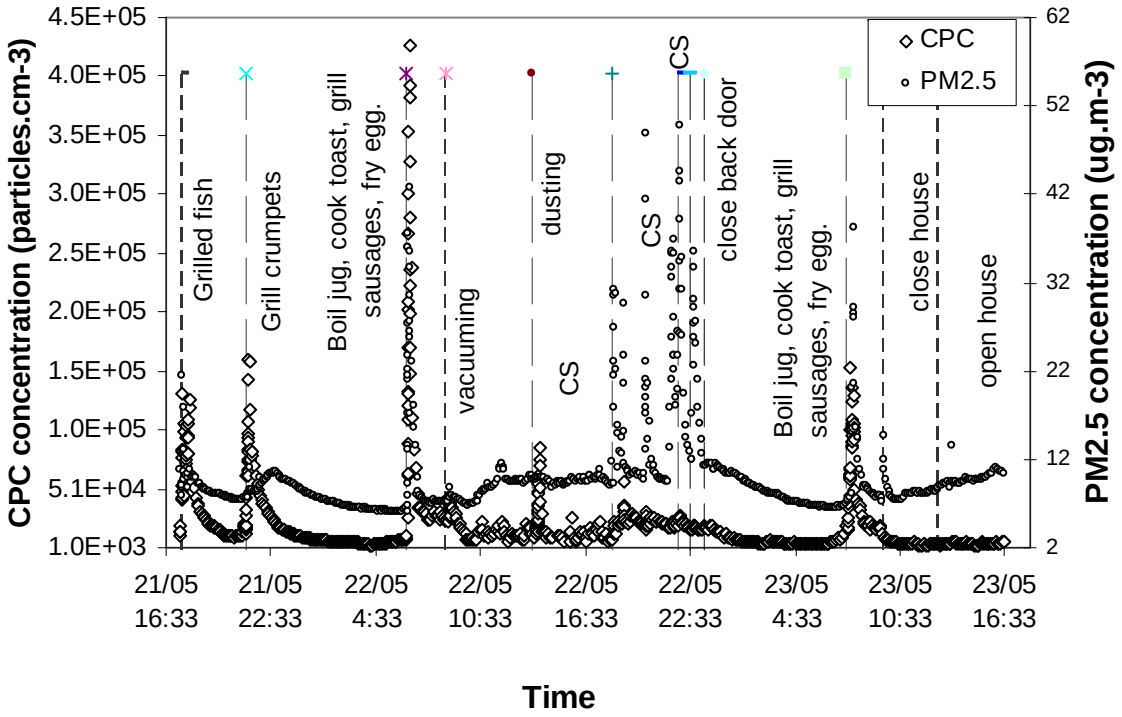


Figure 1. House3, 9-12/July1999, CPC and approximation of PM2.5 concentration in Kitchen, 48h


CPC concentration (particles.cm-3)
4.5E+05
4.0E+05
3.5E+05
3.0E+05
2.5E+05
2.0E+05
1.5E+05
1.0E+05
5.1E+04
1.0E+03
PM2.5 concentration (ug.m-3)
62
52
42
32
22
12
2
CPC
PM2.5
Grilled fish
Grill crumpets
Boil jug, cook toast, grill sausages, fry egg.
vacuuming
dusting
CS
CS
CS
close back door
Boil jug, cook toast, grill sausages, fry egg.
close house
open house
21/05 16:33
21/05 22:33
22/05 4:33
22/05 10:33
22/05 16:33
22/05 22:33
23/05 4:33
23/05 10:33
23/05 16:33
Time

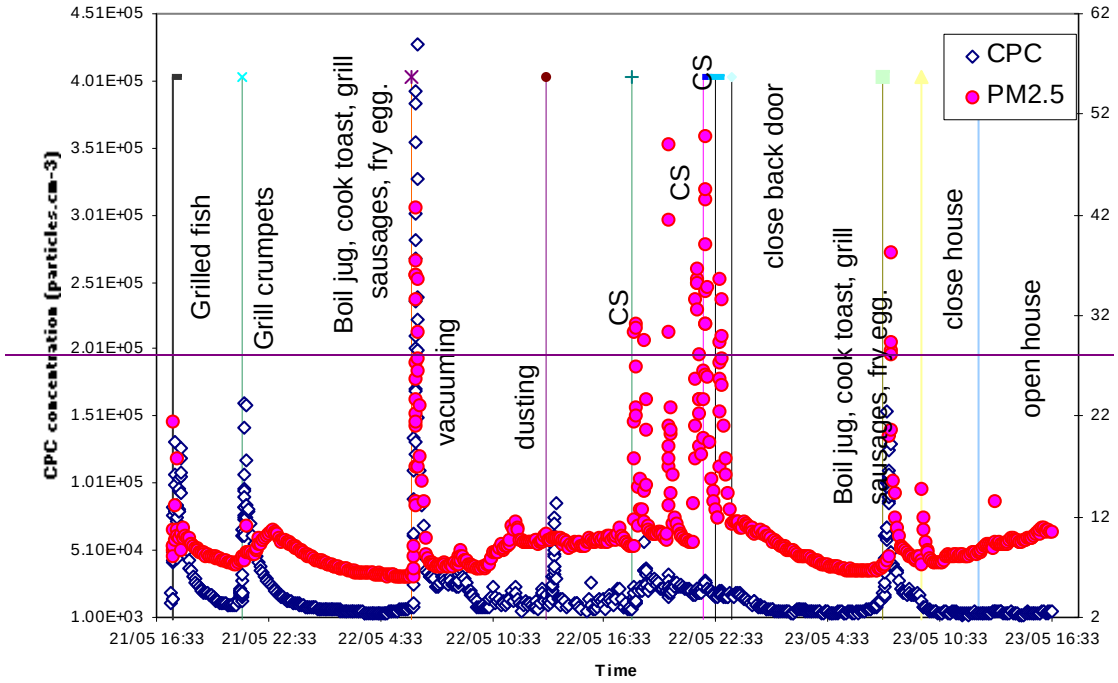


Figure 2. House 12, 21-23/May/1999, CPC and approximation of $PM_{2.5}$ concentrations in kitchen, 48h (CS: cigarette smoking)